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Design and control of a bench-scale reverse-flow reactor for VOC combustion

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Forced unsteady-state gas-solid catalytic reactors have been deeply investigated in the past, as it has been demonstrated that temperature and composition distribution, which cannot be obtained in any steady-state regime, can be obtained by means of forced variations of the inlet parameters, thus enhancing the conversion and selectivity of some chemical processes (1). In particular the periodic reversal of the feed direction allows the autothermal combustion of lean mixtures of waste gases, as the reversal of the flow keeps the heat of reaction inside the bed, thus allowing regenerative heat recovery (2-3). These emissions can be strongly variable in composition and flow rate; as a consequence control measures have to be taken in order to prevent the reactor from extinction during times of lean feed as well as from overheating during times of rich feed. Both problems have been addressed in the past (4), leading to different technical solutions, which are not always feasible or working properly. In this work a new model-based control strategy has been proposed that has been demonstrated to be efficient in controlling both the extinction and the catalyst overheating.

The combustion of lean methane mixtures has been considered by means of numerical simulations and experiments. The simulations were performed using a FORTRAN code that solves the equations corresponding to the mass and heat balances of a heterogeneous and one-dimensional model; plug flow is assumed for the gas phase, with dispersive transport of mass and energy. The transient term is taken into account in the gas phase equations and in the energy equation for the solid phase, while the mass accumulation term is neglected at the solid catalytic surface. As a bench scale unit is employed for validation, an additional energy balance for the reactor wall has been included to take into account radial heat losses from the bed to the wall and axial conduction at the wall: these effects are particularly relevant in reactors with small diameter, such as lab or bench scale units used for research purposes, or small industrial units, while large industrial reactors can be considered to operate adiabatically because of their high bed diameter/wall thickness ratio (5).

The experimental set-up consists of an adiabatic bench scale reactor of 500 mm length and 50 mm diameter; a stainless steel wall has been used, with a special temperature control system based on a dynamic compensation of the thermal losses to get adiabatic behaviour. In order to test the effectiveness of this method and to choose the best configuration (number of heaters and their length) a large number of simulations have been carried out considering the presence of such a device. The experimental results obtained with the bench-scale reactor for the combustion of methane in air using either a palladium and a metal-oxide catalyst supported on alumina confirmed the predictions. Good agreement between the experimental values and the simulations (where real adiabatic behaviour has been considered) are obtained both for the reactor temperature and for the outlet conversion, as it can be seen in Figure 1a. The influence of the main operating parameters has been studied both with simulations and experimentally, thus optimising the reactor performance.

By means of numerical simulation a stability map of the reactor can be obtained (Figure 1b) for a particular value of the inlet flow rate; similar diagrams can be obtained for other values of the flow rate (in the range of feasible values), thus leading to the definition of regions where maximum temperature (or conversion) is higher or lower than a certain value. Because of process constraints on outlet conversion and maximum

temperature on the solid, a well defined operating region is obtained; in Figure 2 the region corresponding to conversion higher than 99.95 % and maximum solid temperatures lower than 650 °C is evidenced.

Until the point corresponding to the operating conditions (inlet concentration and switching period) is in the optimal region no control action is taken; if the inlet concentration increases too much, the control system has two possibilities: it may increase the switching time (as the maximum temperature slightly decreases when the switching time is increased) or, if this is not sufficient, it may dilute the feed. If the inlet concentration decreases too much a control action is taken just when the outlet conversion starts decreasing (because of the thermal inertia of the system, the reactor is able to sustain periods of low inlet concentration, with no decrease in the outlet conversion): either the switching time is reduced, if this can increase the conversion, or auxiliary fuel is added when the inlet concentration is lower than the minimum value that allows adiabatic operation.

In order to apply this control strategy we need to know both the inlet flow rate (and this is easy to do and inexpensive) and the inlet concentration. As on-line measurements may be difficult and expensive, a soft-sensor based on high gain techniques (6) has been used to estimate the inlet concentration from some temperature measurements. The soft-sensor has been demonstrated to give correct prediction of inlet concentration and outlet conversion both when the reactor is fully ignited and when conversion is decreasing.

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Figure

- a) Comparison between experimental (■) and simulated (—) value of solid temperature (on the left) and of the outlet methane concentration (on the right) when the pseudo-steady state has been reached (switching time = 600 s, inlet concentration 2500 ppmV, metal-oxide catalyst).
- b) Example of stability map of a reverse-flow reactor (inlet gas velocity 0.14 m s⁻¹): the continuous line separates the region where extinction occurs from the region where stable operation can be obtained; dotted lines separate the regions where conversion is larger (upper part) or lower than the specified value; dashed lines separate the regions where maximum solid temperature is higher (upper part) or lower than the specified value.

